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Modeled De Facto Reuse and Contaminants of Emerging Concern in Drinking Water Source Waters

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De facto reuse is the percentage of drinking water treatment plant (DWTP) intake potentially composed of effluent discharged from upstream wastewater treatment plants (WWTPs). Results from grab samples and a De Facto Reuse in our Nation's Consumable Supply (DRINCS) geospatial watershed model were used to quantify contaminants of emerging concern (CECs) concentrations at DWTP intakes to qualitatively compare exposure risks obtained by the two approaches. Between nine and 71 CECs were detected in grab samples. The number of upstream WWTP discharges ranged from 0 to >1,000; comparative de facto reuse results from DRINCS ranged from <0.1 to 13% during average flow and >80% during lower streamflows. Correlation between chemicals detected and DRINCS modeling results were observed, particularly DWTPs withdrawing from midsize water bodies. This comparison advances the utility of DRINCS to identify locations of DWTPs for future CEC sampling and treatment technology testing.

Keywords: *contaminants of emerging concern, de facto reuse, drinking water source water, transport and fate*

Raw drinking water supplies are commonly under the influence of treated wastewater discharged upstream of drinking water treatment plant (DWTP) surface water intakes, a situation identified as unplanned or unintentional indirect potable reuse (i.e., de facto reuse [DFR]). For drinking water supplies serving more than 10,000 people from surface water sources, roughly half of these facilities in the United States are affected by at least one upstream treated wastewater discharge, based on previous geographical information systems (GIS)-based modeling efforts (Rice et al. 2015, 2013; Rice & Westerhoff 2015). Concurrently, studies of drinking water sources have detected pharmaceuticals and other contaminants of emerging concern (CECs) of wastewater origin when these sources are located downstream of treated wastewater discharge locations (Bradley et al. 2017, Glassmeyer et al. 2005). Although environmental sampling is the only sure means to identify and quantify contaminants present in a given water body, such monitoring campaigns can be costly. The interpretations of the results, which typically reflect specific conditions at a fixed point in time, are complicated by daily or seasonal differences in wastewater discharge flows or natural hydrologic streamflow variations in the rivers between the points of wastewater discharge and downstream drinking water intake. In theory, tens of thousands of CECs of wastewater origin and their corresponding transformation products could be monitored in water, and multiple preferred lists of approximately 1,000 target or surrogate compounds have been identified as indicators of wastewater in rivers (Bradley et al. 2017; Dickenson et al. 2011, 2009; Mawhinney et al. 2011; Kolpin et al. 2002). Chemical mixture complexity is further amplified when multiple wastewater treatment plants (WWTPs) discharge into the same watershed that provides water to downstream drinking water intakes.

To better understand a drinking water utility's potential contribution to human or ecological exposure to organic CECs of wastewater origin under a range of streamflow conditions, a model (De Facto Reuse in our Nation's Consumable Supply [DRINCS]) has been developed to estimate the DFR across the United States (Rice et al. 2015, 2013; Rice & Westerhoff 2015). Herein we compare results from a specific sampling effort analyzing surface water intakes from 22 surface water treatment plants for 192 organic CECs, with predictions of DFR from DRINCS. The relative location and distance of WWTP discharge points upstream of DWTP intakes are presented, along with the design capacity of the WWTPs, to aid in the comparison and interpretation of the model and chemical results. The objective is to increase the understanding of how the proximity of upstream WWTP discharges increases the vulnerability of downstream surface water DWTPs to contaminants of wastewater origin across the United States.

METHODS

Water treatment plant selection and CEC monitoring information. CEC occurrence information for source water and treated waters at DWTPs were previously reported by Glassmeyer et al. (2017). This study uses the data from Phase II of Glassmeyer et al. (2017) for the 192 organic CECs from the 22 surface water DWTPs.

A detailed description of the criteria used to select sampling sites, sample collection procedure, analysis methods, and quality assurance and control protocols has been previously published (Glassmeyer et al. 2017). In summary, intake grab samples from DWTPs were collected by personnel at participating DWTPs. Samples were packed on ice and shipped overnight to their destination laboratories at the US Environmental Protection Agency (USEPA) or US Geological Survey (USGS) for analysis. All methods have been described previously (Conley et al. 2017; Boone et al. 2014; Furlong et al. 2014, 2008; Batt et al. 2008; Schultz & Furlong 2008; Zaugg et al. 2006; Ternes et al. 2005; USEPA 2005, 2001, 1994; Cahill et al. 2004).

Qualitative versus quantitative detections. Quantitative CEC concentrations and qualitative detection frequencies were previously reported (Glassmeyer et al. 2017). Samples that did not exceed their associated minimum reporting concentration—whether a lowest concentration minimum reporting level or reporting limit—but were above the instrument detection limit were considered a CEC qualitative detection (Glassmeyer et al. 2017). Additionally, samples with associated laboratory fortified matrix samples with >150% recovery were also considered qualitative detections. For both subdetection limit and matrix enhancement scenarios, we were sufficiently uncertain of the actual concentration that we did not report quantitative concentrations. "Qualitative detection frequency" used in the tables and figures in this study includes the detection limit and matrix enhancement censored analytes as well as the quantitatively reported analytes (i.e., every analyte that can be considered a positive detection). Quantitative detection frequency includes only those analyte detections with a concentration that can be reported with analytical certainty.

DRINCS model. The DRINCS model, previously developed and validated by Rice et al. (2015, 2013) and Rice and Westerhoff (2015), is a GIS model that incorporates spatially resolved data layers on the national hydrologic network. The outputs of DRINCS are calculated values of DFR linked with geospatial locations that can be mapped. Discharge locations of treated wastewater from WWTPs and sampling sites of surface water source at DWTPs were used to estimate DFR at drinking water intake under average streamflow conditions. Spatial hydrography data in the United States were obtained from the National Hydrography Dataset Plus (NHD Plus 2012, Viger et al. 2016), which represents the nation's drainage networks and related features, including rivers, streams, canals, lakes, ponds, glaciers, coastlines, dams, and USGS stream gauge data. USGS stream gauge attribute data include average, minimum, maximum, and percentile streamflows. The statistical values were calculated on the basis of the entire record period ending Apr. 20, 2004, which is the end date for the NHD database employed (USEPA 2007).

The WWTP locations and attribute data were obtained from the Clean Watershed Needs Survey 2008 (CWNS 2008), which included 15,837 municipal WWTPs in the United States; we included the facilities (n = 14,651) that currently discharge to surface waters. Supporting attribute data for WWTPs included facility name, National Pollutant Discharge Elimination System (NPDES) permit number, level of treatment (primary, secondary, and tertiary), and present design flow. The level of treatment of effluent at WWTPs is a driver determining the potential occurrence of CECs, as concentrations substantially decrease with higher levels of treatment. Our analysis of treated municipal wastewater discharges from WWTPs included combined sewer systems but did not take into consideration combined sewer overflows or wet-weather bypasses (both of which can yield significant CEC loads; Phillips et al. 2012), or non-WWTP entities with NPDES permits to discharge. Conservative, and possibly worst-case, assumptions in calculating DFR made in the previous study (Rice et al. 2013) were used and include (1) WWTP discharge was equal to that of the present design capacity; (2) WWTP effluent had no in-stream loss; and (3) all water bodies were completely mixed. A python program automated the process performed in the previous study (Rice et al. 2013). Levels of DFR were calculated from the cumulative upstream WWTP design discharges ($\Sigma Q_{ww,i}$) divided by the streamflow at the surface water DWTP intake (Q_{SW}):

$$\mathrm{DFR} = \frac{\sum Q_{ww,i}}{Q_{SW}} \times 100\% \tag{1}$$

Network relationships in streams between upstream WWTPs and each receiving DWTP at a regional scale were derived using a computerized mapping and analytics platform.[1] Flow direction was established using the digitized direction from an attribute table of stream network (NHD). The tracked upstream streamflow obtained from the geometric network was used to build a network analysis of routes (waterways). Using network analyst tools, proximal river distances from WWTPs upstream to each DWTP were determined by using the closest facility function based on Dijkstra's algorithm (Allen 2013, Dijkstra 1959). Before use at all of the DWTP intakes, the Dijkstra algorithm approach was validated against the Ruler tool in Google Earth.

RESULTS

Detection of CECs at DWTP intakes. As previously reported (Glassmeyer et al. 2017), the number of qualitatively detected analytes in the source water ranged from 30 in DWTP 29 to a maximum of 104 in DWTP 4. Excluding detection frequency for microbial and inorganic chemicals we are not considering in this analysis, the number of the 192 organic CECs qualitatively detected ranged from nine in DWTP 5 to 71 in DWTP 4.

Magnitude and factors influencing DFR levels at DWTPs *DFR under mean streamflow.* The DRINCS predicted levels of DFR ranged from 0 to 12.8% under mean streamflow (Table 1). Three DWTPs (13, 23, and 29) on surface waters had no predicted wastewater impacts, as there were no upstream WWTP discharges identified within the watershed. Three DWTPs (5, 12, and 24) were groundwater systems, and DFR values were not predicted for these. Nineteen of the 22 DWTPs in Phase II with surface water sources had at least one upstream WWTP discharge, and DFR levels could be predicted by DRINCS. These DWTPs will be the focus in the remaining part of this section.

Streamflow and Strahler stream order. Impacts of varying streamflow (daily, seasonal, and annual) were considered two ways in the DRINCS model. First, historical streamflow data were used to obtain fifth and 90th percentile streamflows because these influence the potential range of higher to lower DFR values, respectively, that could be expected to occur at a DWTP intake. Second, source waters were classified on the basis of Strahler stream order (Strahler 1957). For this classification, each segment of a river within a watershed can be considered as a node in a tree. When two first-order streams come together, they form a second-order stream, two second-order streams must flow together to form a third-order stream, and so on. Streams of lower order joining a higher-order stream do not change the order of the higher stream. With two exceptions, the surface water DWTPs in this study are located on fifth- to ninth-order streams (Table 1).

Figure 1 summarizes these ranges in streamflow (part A) and associated DFR for DWTPs (part B), further classified based upon Strahler stream order. Generally, higher stream orders have higher mean flow rates (Figure 1, part A). The ratio of 90th:5th percentile streamflows (Q_{90}/Q_5) provides a relative indicator for the potential variation in streamflow. Table 1 summarizes Q_{90}/Q_5 ratio values, which range from 4 to 36. Higher streamflow dilutes wastewater discharged upstream of DWTP intakes. Variations in streamflow are generally seasonal and larger than variations in discharge flow rate from WWTPs, which fluctuate diurnally and are designed with peak hourly discharge to average daily discharge ratios of between 2 and 4. Whereas wet-weather wastewater discharges can be quite variable, the difference between winter and summer wastewater discharge flow rates is generally less than a factor of 2. Thus, variations in streamflow would be expected to alter DFR (Eq 1) to a larger extent than variations in wastewater discharge flow rates.

DFR values are shown in Figure 1, part B, on a log scale, with the open circle representing DFR at median streamflow; DFR at mean streamflow is listed in Table 1. Higher DFR values occur at lower streamflows (Eq 1). Although exceptions occur, higher Strahler stream orders have lower DFR values (Figure 1, part B). The highest predicted DFR under low flow conditions is 84% at DWTP 4 (Figure 1, part B); mean DFR is listed in Table 1. The highest maximum DFR values are for DWTPs located on fifth- and sixth-order streams (Figure 1, part B).

Number and size of upstream WWTP discharges. DWTPs experience different amounts of DFR based on the presence of upstream WWTPs. Table 2 provides information on the number and design treatment capacity of WWTPs located upstream of each DWTP intake. DWTP 2 and 21 (plants nearly co-located on the same river) have 1,200 upstream WWTPs that account for up to 1,372 mgd of treated wastewater, but 442 of these WWTPs are small and have design discharges below 0.1 mgd. At an average daily sewage production of 75 gpd per person, a 0.1 mgd facility serves a population of roughly 1,300 people. In total, 3,615 (82%) out of 4,392 of the WWTPs upstream of the DWTPs in this study have design capacities below 1 mgd (Table 2).

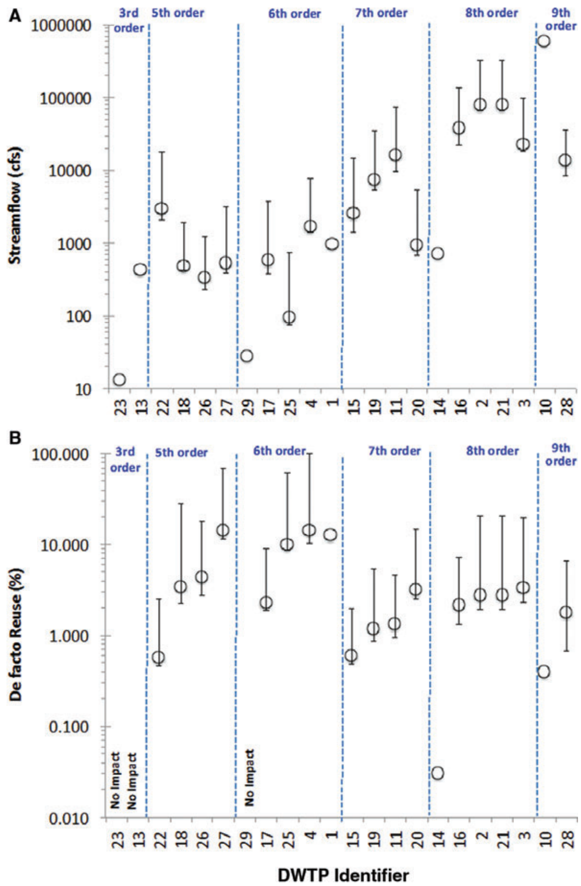
TABLE 1 Geographical and hydrological information on sampled locations

DWTP #	Region of the United States	Water Source	Stream Order	Q90: Q5 Ratio	Reported Relative Water Level at Sampling	Annual Mean Streamflow at DWTP Intake *cfs*	De Facto Reuse at Mean Streamflow %
DWTP 1	South Central	River	6	NA	Average	966	12.8
DWTP 2	North Central	River	8	21	Slightly low	95,353	2.2
DWTP 3	North East	River	8	16	Average	32,861	2.3
DWTP 4	North East	River	6	21	Average	3,026	7.8
DWTP 5	South West	Groundwater	—	—	—	—	—
DWTP 10	South Central	River	9	NA	Low	587,823	0.4
DWTP 11	North West	River	7	9	Above average	27,399	0.8
DWTP 12	North Central	Groundwater	—	—	—	—	—
DWTP 13	North East	Reservoir	3	12.3	Average	419	0
DWTP 14	South East	Lake	8	NA	Average	718	0.03
DWTP 15	Plains	River	7	11	Average	4,498	0.3
DWTP 16	Plains	River	8	6	High	30,980	2.7
DWTP 17	South East	River	6	15	Above average	1,056	1.3
DWTP 18	North East	River	5	20	Average	644	2.6
DWTP 19	Plains	River	7	14	Average	8,581	1.0
DWTP 20	South West	River	7	16	Average	2,006	1.5
DWTP 21	North Central	River	8	21	Average	95,353	2.2
DWTP 22	South East	River	5	16	Average	1,817	0.9
DWTP 23	South East	Reservoir	3	13	Low	16	0
DWTP 24	Plains	Groundwater	—	—	—	—	—
DWTP 25	Plains	Reservoir	6	36	Very low	213	4.3
DWTP 26	North East	River	5	8	Average	527	2.8
DWTP 27	North Central	River	5	19	Low	1,177	6.4
DWTP 28	South West	Reservoir	9	4	Low	16,635	1.4
DWTP 29	South Central	Reservoir	6	NA	Average	28	0

DWTP—drinking water treatment plant, NA–not applicable, Q90:Q5–the ratio of 90th:5th percentile streamflows

Dashed lines indicate groundwater locations therefore excluded from calculation.

FIGURE 1 Median and variations in streamflow (A) and de facto reuse (DFR) percentage values (B) for DWTPs



DWTP—drinking water treatment plant

Circles represent the median. Variations (fifth or 90th percentile values) are shown in whiskers. DWTPs are grouped by Strahler stream order classification.

Only 59 out of nearly 4,392 WWTPs considered in this study have design capacities >10 mgd (Table 2). Thus, there are large numbers of small WWTPs in the studied watersheds.

Proximity of upstream WWTP discharges. DFR values were calculated from the cumulative upstream wastewater discharges (Eq 1), and do not directly account for the proximity between WWTP discharges and the DWTP intake. A single value to represent proximity was difficult to derive because of the complexity of watersheds. Figure 2 illustrates watersheds and location of WWTP discharges upstream of four DWTP intakes (DWTPs 3, 4, 15, and 16). There are complex networks of tributaries in these watersheds, many with several WWTP discharges. Figure 3 documents the proximal distance between upstream WWTP discharges and the DWTP intake for several watersheds; each symbol represents a WWTP discharge. The y-axis in Figure 3 represents the distribution of treated wastewater flows from upstream WWTPs ($Q_{ww,i}$), normalized to cumulative wastewater flow from all upstream WWTPs ($Q_{ww,T}$), separated by stream order. The value of $Q_{ww,T}$ varies by site and is summarized in the last column of Table 2. Eleven of the DWTPs (e.g., DWTPs 18 and 26 in Figure 3, part A) have WWTP discharges located within 10 mi upstream of a DWTP intake. In others, few individual WWTPs contribute substantially to the overall wastewater flows into the surface water source serving the DWTP, as indicated by breaks or jumps in the plot (e.g., DWTP 19 in Figure 3, part C, and DWTP 3 in Figure 3, part D). In other systems (e.g., DWTPs 2 and 16 in Figure 3, part D), most of the wastewater flow originates hundreds of miles upstream.

Several indictors were used to quantify the complex relationship between multiple WWTP discharge points located at multiple upstream locations. As illustrated in Figure 3, any DWTP location can be modeled as having a distribution of upstream sources. Several statistical functions can be used to fit such distributions and serve as indicators, but they typically require multiple fitted parameters. As discussed subsequently, the purpose of such indicators is to provide a secondary index of relative risk of having CECs of wastewater origin in DWTP source supplies. Consequently, correlating multiple fitted parameters were not deemed appropriate. Two alternative indexes were developed to further examine the relation between WWTP locations and CEC detections. To calculate the first index, the magnitude ($Q_{ww,i}$ in mgd) and distance (M_i in mi) of each (i) WWTP located upstream were considered to be inversely proportional (Eq 2) to relative risk of CEC occurrence in DWTP intakes; $Q_{WW,T}$ is the cumulative discharge of all upstream WWTPs ($Q_{WW,T} = \Sigma Q_{WW,i}$). A singular proximity index (PI) from all upstream WWTPs was then calculated by summing these values (Eq 3). Larger PI values suggest larger WWTP discharges located closer to DWTP intakes, and could indicate a larger potential wastewater impact. For the second index, the relative skewness (SK) of the distribution functions illustrated in Figure 3 (F being the y-axis distribution from 0 to 1) were considered as potentially being useful indicators for differences among watersheds. A simple metric was used to quantify this SK. As shown in Eq 4, SK was related to the distance ($M_{0.1}$) associated with $F = 0.1$ divided by the distance ($M_{0.5}$) associated with $F = 0.5$ in Figure 3; F is the normalized cumulative distributions of WWTP wastewater flows (i.e., y-axis value in Figure 3). SK would range from 0 to 1. Higher SK values suggest larger WWTPs located closer to the DWTP. There is no direct relation between PI and SK, but each index can be used separately to compare

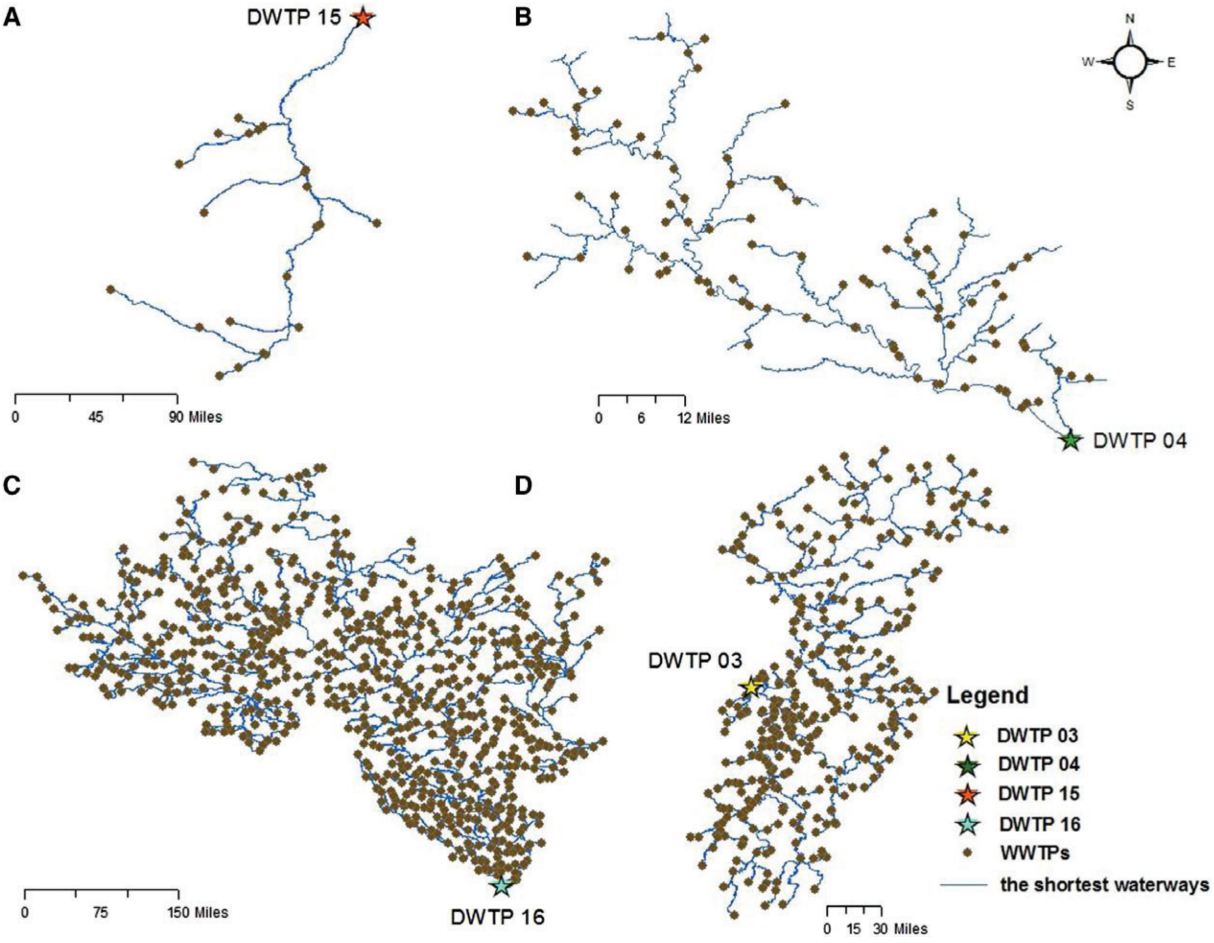
TABLE 2 Location and sizes of wastewater treatment plants located upstream of drinking water treatment plants investigated in this study

	Location of Upstream WWTPs		Size of Upstream WWTPs				Total Impact of Upstream WWTPs	
DWTP #	Less than 10 mi *n*	Maximum distance *mi*	Less than 0.1 mgd *n*	0.1–1 mgd *n*	1–10 mgd *n*	Greater than 10 mgd *n*	Total Upstream WWTPs *n*	Accumulated Upstream WW Discharges $Q_{ww,T}$ *mgd*
DWTP 1	0	115	4	14	11	2	31	80
DWTP 2	1	771	442	529	215	14	1,200	1,372
DWTP 3	1	315	103	171	73	3	350	491
DWTP 4	3	116	16	42	34	1	93	153
DWTP 5	—	—	—	—	—	—	—	—
DWTP 10	6	905	178	212	67	8	465	1,459
DWTP 11	0	187	4	27	8	3	42	135
DWTP 12	—	—	—	—	—	—	—	—
DWTP 13	0	0	0	0	0	0	0	0
DWTP 14	0	54	0	1	0	0	1	0.15
DWTP 15	0	384	10	9	3	0	22	10
DWTP 16	3	785	388	326	60	9	783	534
DWTP 17	1	63	0	2	2	0	4	9
DWTP 18	1	38	0	4	2	0	6	11
DWTP 19	0	386	51	62	10	1	124	56
DWTP 20	0	306	7	11	5	0	23	19
DWTP 21	1	771	442	529	215	14	1,200	1,372
DWTP 22	1	126	3	6	2	0	11	11
DWTP 23	0	0	0	0	0	0	0	0
DWTP 24	—	—	—	—	—	—	—	—
DWTP 25	1	438	0	4	2	0	6	6
DWTP 26	0	45	0	4	2	0	6	9
DWTP 27	1	90	1	10	5	2	18	49
DWTP 28	0	660	1	2	2	2	7	155
DWTP 29	0	0	0	0	0	0	0	0

DWTP—drinking water treatment plant, WW—wastewater, WWTP—wastewater treatment plant

Dashes indicate groundwater locations therefore excluded from calculation.

FIGURE 2 Illustrations of watersheds for four drinking water treatment plants (DWTPs) showing rivers (blue lines), location of multiple wastewater treatment plant (WWTP) discharge locations (solid circles), and locations of DWTPs (star symbols) that represent the downstream end (lower elevation) of the watersheds



proximity patterns among different DWTPs. PI and SK values are summarized in Table 3.

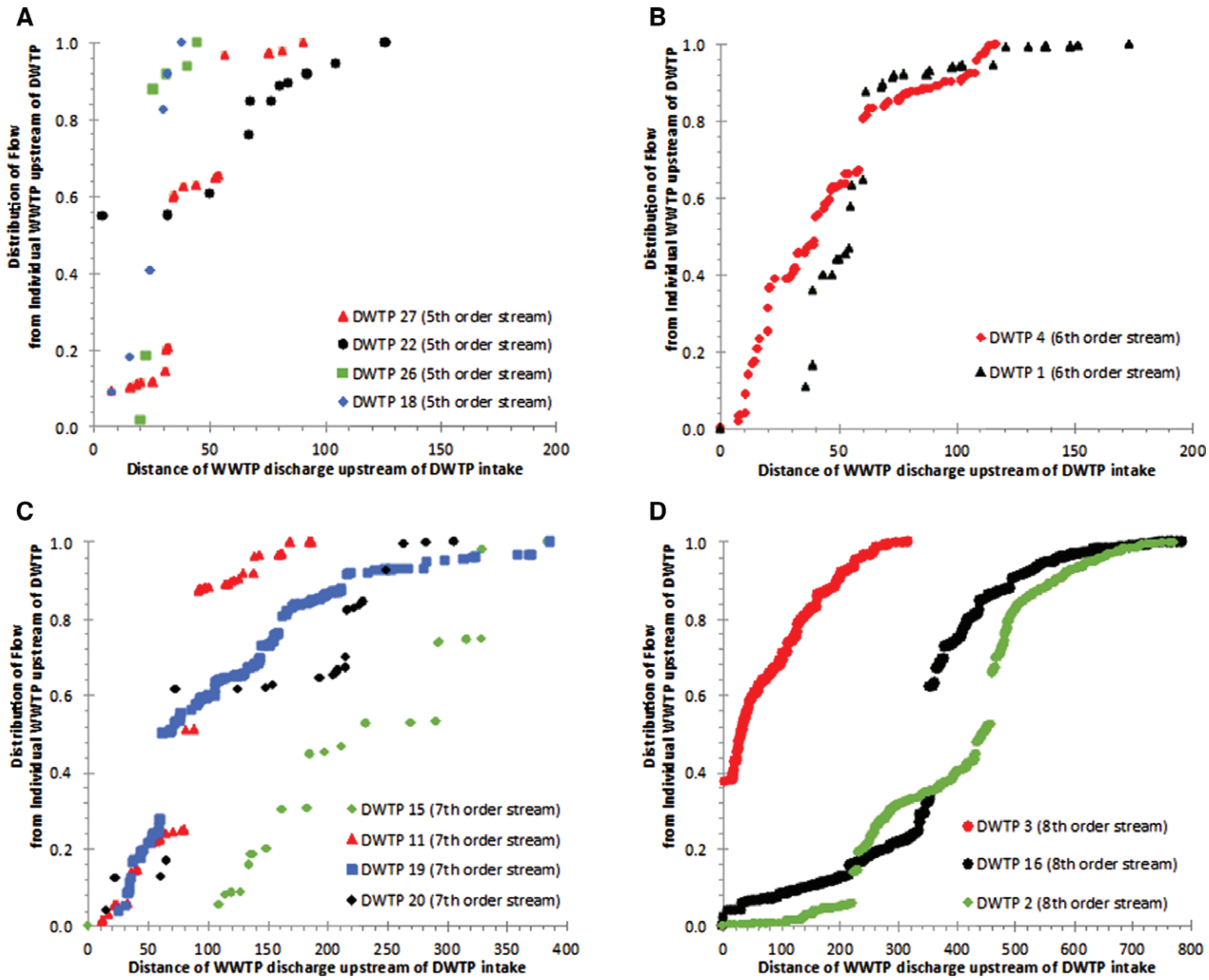
$$PI_i = \frac{Q_{ww,i}}{M_i} \tag{2}$$

$$\frac{PI = \sum {}^{Q_{ww,i}}/_{M_i}}{Q_{WW,T} \times 1{,}000} \tag{3}$$

$$SK = {}^{M_{0.1}}/_{M_{0.5}} \tag{4}$$

Qualitative comparison of CEC detection and DRINCS model DFR predictions. The over 4,000 WWTPs present in the watersheds of the DWTPs studied herein include a wide range of treatment processes from aerated lagoons to advanced nutrient control. Biodegradation, biosorption, volatilization, hydrolysis, oxidation, and other biochemical or physical processes within different types of WWTPs can potentially influence the extent of CEC removal. DRINCS does not directly account for these differences in treatment process, and DFR simply represents a conservative estimate for potential risk of having surface DWTP supplies containing CECs of wastewater origin. However, the variability of WWTP unit process upon CEC removal is expected to affect absolute CEC concentrations present at downstream DWTP intakes. The flow of the water body affects the transport time for WWTP effluents to reach the downstream DWTPs. At a velocity of 1 ft/s (0.3 m/s), the travel time is approximately one week for 100 mi; above 10 ft/s (3 m/s), the travel time is less than a day—one day for 100 mi. CECs can continue to biodegrade in water after wastewater is discharged to rivers and before entering DWTPs. Additional biogeochemical processes (adsorption to sediment, volatilization, photolysis, and so on) can also occur in rivers (Chen

FIGURE 3 Normalized cumulative distributions (*F*, dimensionless) of WWTP wastewater flows, relative to the total upstream WWTP flow



DWTP—drinking water treatment plant WWTP—wastewater treatment plant

Each point represents a WWTP discharge.

et al. 2010, 2009, 2008a, 2008b). These time-dependent processes depend on other water quality factors (temperature, pH, turbidity, and so on), location, river depth, and others. The CECs within this data set will have a range of persistence both during wastewater treatment and environmental transport (Glassmeyer et al. 2005). In general, the per- and polyfluoroalkyl substances (PFASs) are more resistant to treatment (Zhang et al. 2015, Rahman et al. 2014) and more stable in the environment (Nguyen et al. 2017, Happonen et al. 2016, Wang et al. 2015) than most CECs.

The CEC source water data set is comprehensive both in terms of the number of chemicals analyzed and number of DWTPs sampled (n = 25). However, grab samples are only representative of a single point in time, many of the CECs were below reporting or detection limits, and quantitative concentrations were not reported. Therefore, the researchers made a qualitative comparison of CEC occurrence and DRINCS model outputs rather than using a formal statistical analysis. Table 4 provides a cursory comparison of the field and modeling potential for CECs to occur at DWTP intakes. The general trend is that more CECs are detected at higher DFR values. Figure 4 displays the number of qualitative detections for all of the CECs at each location. In general, within each stream order, the number of detected analytes increases as DFR increases. One notable exception to this trend is DWTP 1. The field blanks associated with this location had measurable concentrations of many commonly detected analytes,

TABLE 3 Results of modeling and summary of chemical analyses of drinking water source water samples

DWTP #	De Facto Reuse at Mean Streamflow %	Proximity Index (PI)	Skewness Index (SK)	Number of Organic Chemicals Qualitatively Detected (*n* = 192)	Number of Organic Chemicals Quantitatively Detected (*n* = 192)	Sum Quantitative Detections *ng/L*
DWTP 1	12.8	20	0.67	31	16	135.3
DWTP 2	2.2	3.5	0.57	32	18	1,075.2
DWTP 3	2.3	169	1.0	55	31	820.2
DWTP 4	7.8	40	0.26	71	35	1,425.5
DWTP 5	—	—	—	9	4	27.5
DWTP 10	0.4	43	0.02	28	18	460.9
DWTP 11	0.8	16	0.41	15	6	1.5
DWTP 12	—	—	—	18	11	69.9
DWTP 13	0	0	0	12	9	3.3
DWTP 14	0.03	19	1.00	13	8	2.7
DWTP 15	0.3	4.9	0.69	14	6	1.5
DWTP 16	2.7	30	0.42	15	13	350.1
DWTP 17	1.3	39	1.00	26	13	293.4
DWTP 18	2.6	46	0.24	26	22	265.6
DWTP 19	1.0	15	0.50	24	10	27.6
DWTP 20	1.5	15	0.31	22	8	20.0
DWTP 21	2.2	3.5	0.86	36	20	877.6
DWTP 22	0.9	156	1.00	37	26	1,762.2
DWTP 23	0	0	0	18	11	138.3
DWTP 24	—	—	—	13	11	541.7
DWTP 25	4.3	64	1.00	19	11	17.7
DWTP 26	2.8	39	0.84	44	20	451.9
DWTP 27	6.4	35	0.44	41	24	1,699.7
DWTP 28	1.4	39	1.00	24	15	232.4
DWTP 29	0	0	0	15	8	31.1

Concentration data source: Glassmeyer et al. 2017.

DWTP—drinking water treatment plant

Dashes indicate groundwater locations therefore excluded from calculation.

TABLE 4 Comparison between de facto reuse (DFR) and number of contaminants of emerging concern (CECs) qualitatively detected at drinking water treatment plant (DWTP) intakes[a]

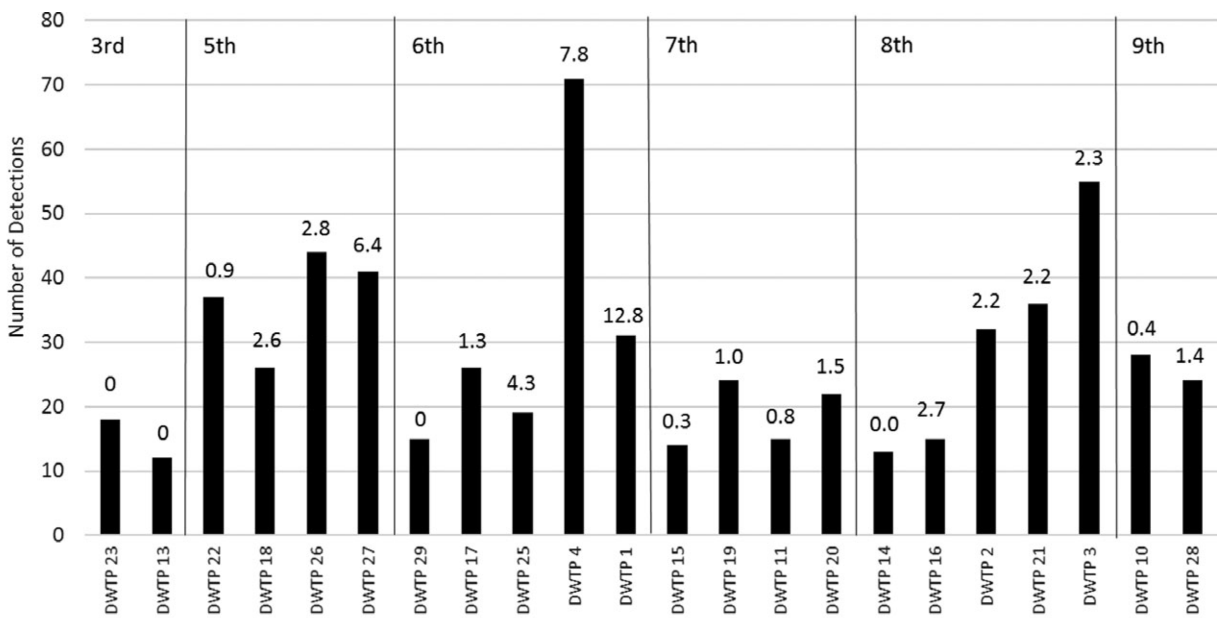
No. of Qualitative Detections of CECs at DWTP Intakes	Level of DFR Determined Under Mean Flow in DRINCS				
	Not Impacted	<0.1%	0.1–1%	1–5%	>5%
<10	—	—	—	—	—
10–20	2	1	2	3	—
20–30	—	—	1	5	—
30–40	—	—	1	2	1
>40	—	—	—	2	2

DRINCS—De Facto Reuse in our Nation's Consumable Supply

[a]Values in the table represent the number of DWTPs in each category.

Dashes indicate groundwater locations therefore excluded from calculation.

FIGURE 4 Number of organic contaminant of emerging concern analytes detected at each sampled location, sorted by stream order



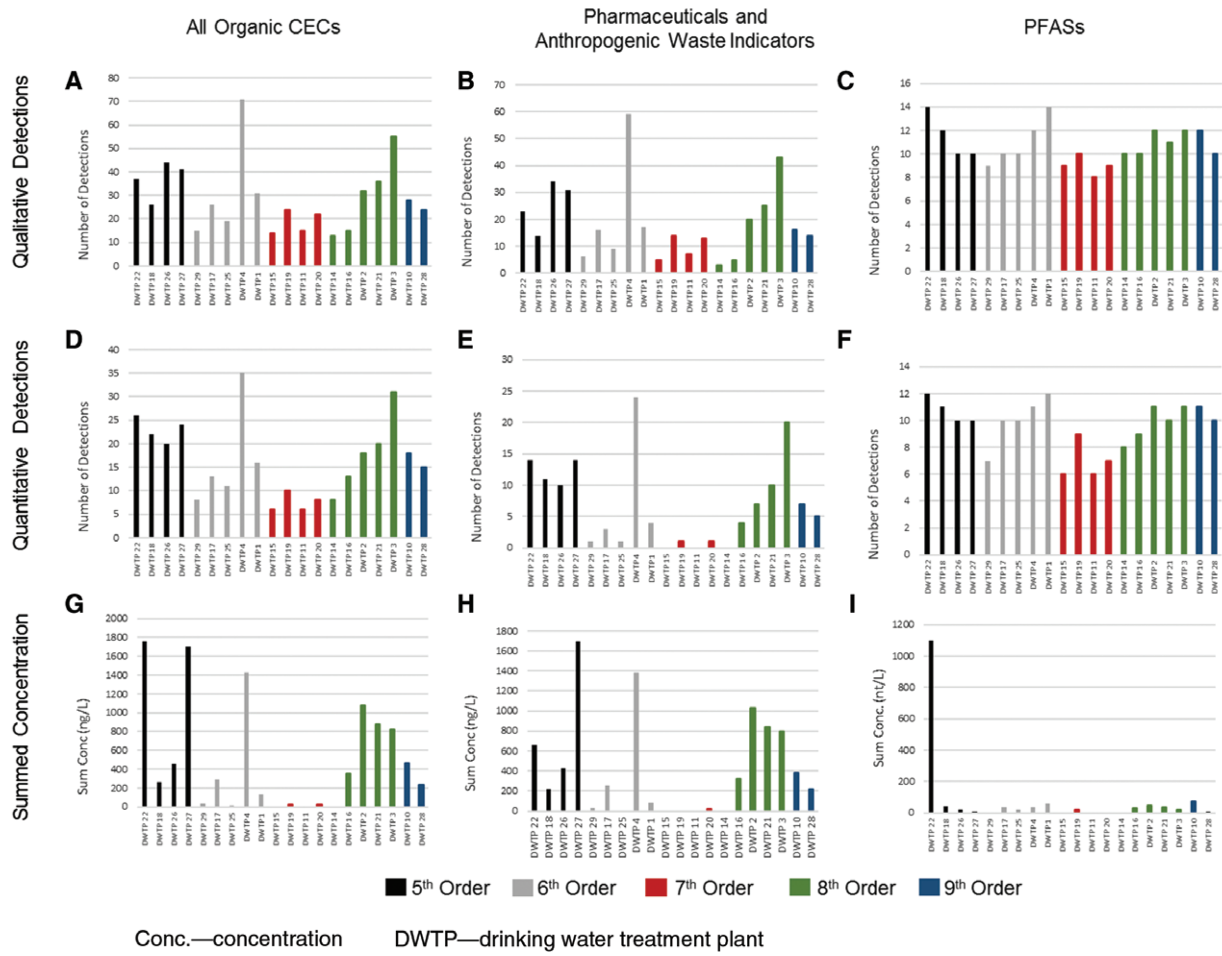
DWTP—drinking water treatment plant

The order of the DWTPs reflects the stream order and de facto reuse (DFR) percentage at median streamflow from Figure 1. The vertical lines on the graph separate the stream orders. The number above each bar is the DFR at mean streamflow (Table 1).

such as atrazine, caffeine, cotinine, meprobamate, coprostanol, galaxolide, *N*,*N*-diethyl-*meta*-toluamide, tri(2-butoxyethyl) phosphate, and tri(2-chloroethyl) phosphate. Per our quality assurance/quality control protocol (Glassmeyer et al. 2017), the detections of these analytes in the DWTP 1 samples were censored, as the concentrations in the samples needed to exceed blank detections by a factor of three or more to be retained. Without the removal of the field blank censored detections, DWTP 1 would have more measured detections, and a general trend of increasing detections with increasing DFR within a stream order would hold.

Figure 5 explores the trends between measured detections and stream order, and Figure 6 examines the relation between concentration and DFR (parts A–C), PI (parts D–F), and SK (parts G–I). This figure examines

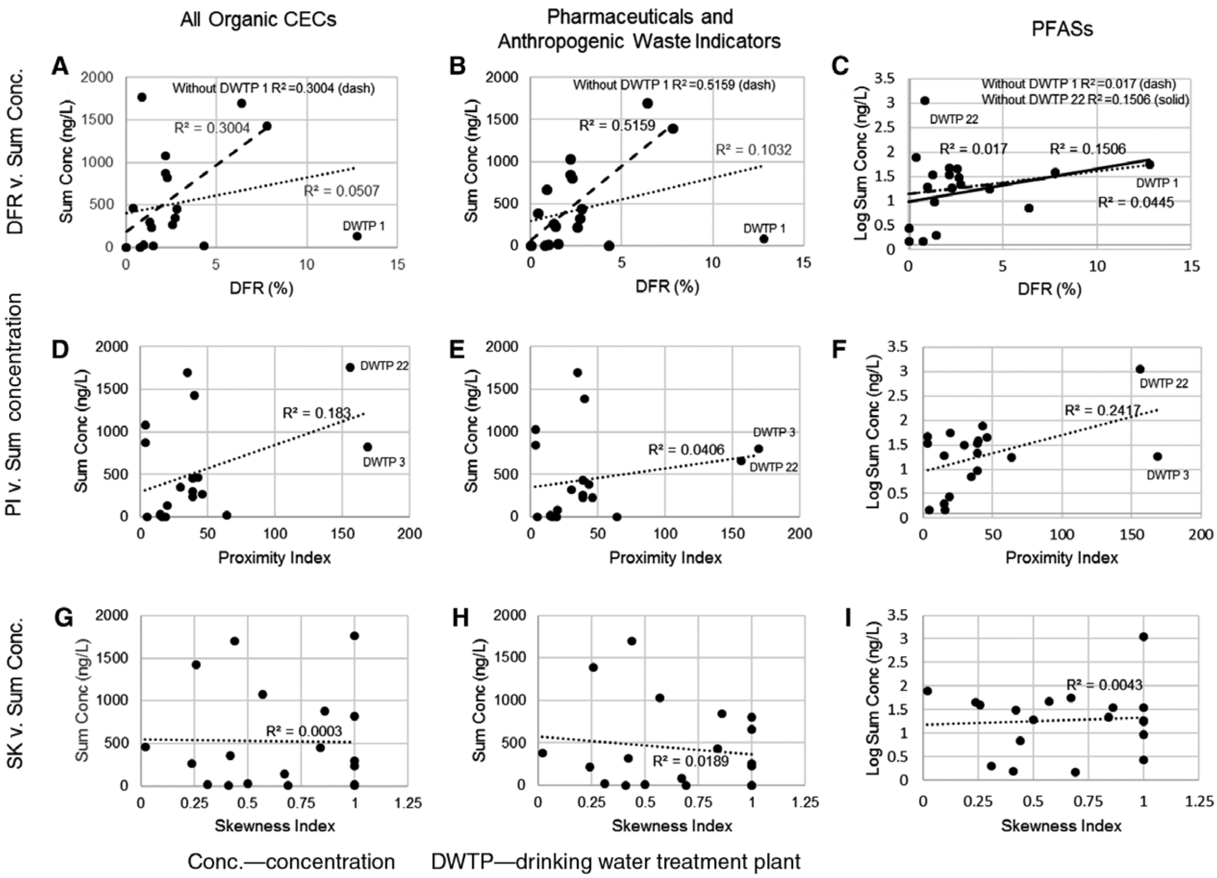
FIGURE 5 Qualitative detections, quantitative detections, and summed concentration for all organic contaminants of emerging concern (CECs), separated by pharmaceuticals and anthropogenic waste indicators and per- and polyfluoroalkyl substances (PFASs)



Bar color separates the DWTPs by Strahler stream order, and they are ordered by de facto reuse (DFR) at median streamflow (Figure 1).

all of the organic CECs (for both Figures 5 and 6, parts A, D, and G) and separates the relatively ephemeral pharmaceuticals and anthropogenic waste indicators (AWIs) (parts B, E, and H) from the more persistent PFASs (parts C, F, and I). In terms of both the qualitative and quantitative detections (Figure 5, parts A–F), the PFASs (parts C and F) show substantially less variability between the locations than the combined pharmaceuticals and AWIs (parts B and E). It is interesting to note that once the PFASs are removed, the DWTPs with seventh-order stream sources tend to have lower numbers of detections than the sixth- or eighth-order sources (Figure 5, parts B and E). More research would be needed to determine if this is a nationwide trend or unique to this data set. The cross-site similarity of the PFASs is diminished when concentration is considered (Figure 5, part I). DWTP 22 had a greater total PFAS concentration compared with the other locations. This illustrates one of the weaknesses of the DRINCS model: although the wastewater composition of the source water is in general a good indicator for relative contamination, unique nonwastewater sources may be significant contributors of CECs upstream of source water intakes. Turning to the three indexes discussed earlier (DFR, PI, and SK; Figure 6), several relationships can be noted. Excluding the DWTP 1 outlier, DFR shows the strongest trend of the three indexes when plotted against the sum of concentrations for all organic CECs (Figure 6, part A) and pharmaceuticals and AWIs (part B); higher DFRs are generally correlated with greater concentrations of CECs in the source water. When DWTP 1 is excluded from the regression, the R^2 increases from 0.0507 to 0.3004 for all organic CECs (Figure 6, part A), and from 0.1032 to 0.5159 for the

FIGURE 6 De facto reuse (DFR) at mean streamflow, proximity index (PI), and skewness (SK) index in relation to the summed concentration for all organic contaminants of emerging concern (CECs), and separated by pharmaceuticals and anthropogenic waste indicators and per- and polylfluroalkyl substances (PFASs)



pharmaceuticals and AWIs (part B). For the PFASs, the trend is minimized by the high concentrations at DWTP 22 (R^2 = 0.0445; Figure 6, part C), but even when that point is omitted, the trend is not as strong as it is for the other analytes (R^2 = 0.1506). Removing the DWTP 1 outlier similarly does not increase the relation (R^2 = 0.017). When PI is plotted against the sum of the three different concentration sets (Figure 6, parts D, E, and F), the trends (R^2 of 0.183, 0.0406, and 0.2417, respectively) are not as strong as DFR trends with the DWTP 1 outlier removed. DWTPs 3 and 22 are two points outside the rest of the field on all the organic CEC and pharmaceutical and AWI-only graphs (and are indicated on Figure 6, parts D and E). Both of these locations have a large percentage of their total wastewater load (~38 and 55%, respectively; Figure 3, parts D and A) from WWTPs within a 10 mi distance. The PI may have application for determining DWTPs that have relatively stronger impacts from individual nearby WWTPs, whereas DFR is a better indicator of the general wastewater impact. The SK value shows no relationship under any of the organic CEC permutations (Figure 6, parts G, H, and I).

To further examine the effect DFR may have on analyte detections, Table 5 compiles the maximum concentration and DWTP with the maximum detection for all 62 organic CECs quantitatively detected at least once in the source water. Eleven of the 25 DWTPs had at least one maximum concentration for any analyte measured in its respective source water, but DWTP 4 is distinctive in this group. It has more than twice as many maximum concentration detections as any other DWTP (with the exception of DWTP 22 PFAS concentrations). Additionally, the detections at DWTP 4 are often the study maximum for organic CECs detected at many locations (as indicated by a >4% quantitative frequency); the other DWTPs often are the maximum detection because they are the only detection. DWTP 4 had the greatest DFR of the 11 DWTPs on this list at 7.8% (Table 3). To see how these maximum concentrations in source water compare with measured concentrations in wastewater-influenced locations, the

TABLE 5 Quantitative detections assessment

Location With Max Detection	Analyte	CAS Number	*n*	Qualitative Frequency of Detection %[a]	Quantitative Frequency of Detection %[a]	Max Source Water Conc *ng/L*[a]	Max WWTP-Influenced Conc *ng/L*[b]	Source as % of WWTP Impacted Conc
DWTP 2	Tri(2-butoxyethyl) phosphate	78-51-3	25	36	4	470.0	—	—
	Tri(2-chloroethyl) phosphate	115-96-8	25	32	4	65.0	—	—
DWTP 3	Tramadol	27203-92-5	25	32	16	1,723.0	1,311.3	1.8
	Trimethoprim	738-70-5	25	28	16	9.9	198.8	5.0
	Diltiazem	42399-41-7	25	20	8	15.5	56.0	27.7
	Ibuprofen	15687-27-1	25	8	8	17.7	1,620.0	1.1
	Furosemide	54-31-9	25	4	4	17.5	—	—
	Paraxanthine	611-59-6	25	4	4	29.2	—	—
DWTP 4	Galaxolide (HHCB)	1222-05-5	25	36	36	110.0	1,400.0	7.9
	Metoprolol	51384-51-1	25	52	32	37.8	367.1	10.3
	Carbamazepine	298-46-4	25	56	28	35.7	382.7	9.3
	Estrone	53-16-7	25	52	20	0.3	31.5	0.9
	Hydrochlorthiazide	58-93-5	25	24	20	67.3	—	—
	Desvenlafaxine	93413-62-8	25	28	16	60.4	1,953.5	3.1
	Bromoform	75-25-2	25	12	12	88.0	—	—
	Caffeine	58-08-2	25	32	12	2,790.9	1,275.9	7.1
	Triclosan	3380-34-5	25	52	12	3.5	534.0	0.7
	Valsartan	137862-53-4	25	20	12	79.2	—	—
	Cotinine	486-56-6	25	16	8	18.9	68.1	27.7
	Fexofenadine	83799-24-0	25	12	8	163.1	2,047.4	8.0
	Venlafaxine	93413-69-5	25	12	8	26.3	407.3	6.5
	Atenolol	29122-68-7	25	28	4	29.8	551.1	5.4
	Diphenhydramine	58-73-1	25	16	4	10.3	145.3	7.1
	Progesterone	57-83-0	25	12	4	0.1	0.9	16.0
DWTP 12	Bisphenol A	80-05-7	25	4	4	28.5	163.0	17.5
DWTP 18	Triclocarban	101-20-2	21	57	24	2.9	—	—
	Dihydrotestosterone	521-18-6	24	4	4	0.3	1.6	19.8
	N,N-Diethyl-*meta*-toluamide	134-62-3	25	48	4	98.0	—	—
	Testosterone	58-22-0	25	4	4	0.2	1.1	14.1
DWTP 21	Atrazine	1912-24-9	25	44	24	323.3	5,170.0	6.3
	Metolachlor	51218-45-2	25	36	12	130.0	1,490.0	8.7
	Norverapamil	67018-85-3	25	20	8	47.2	—	—
	10-hydroxy-amitriptyline	1159-82-6	25	4	4	0.3	9.4	3.2
	Amitriptyline	50-48-6	25	4	4	12.1	10.2	118.1
	Verapamil	52-53-9	25	20	4	45.9	12.9	355.7
DWTP 22	PFHpA	375-85-9	25	96	96	184.0	—	—
	PFHxA	307-24-4	25	96	96	55.1	—	—
	PFNA	375-95-1	25	96	96	41.4	—	—
	PFBA	375-22-4	25	92	92	96.8	—	—
	PFPeA	2706-90-3	25	92	92	501.0	—	—
	PFOS	1763-23-1	25	96	88	48.3	—	—

(Continues)

TABLE 5 Quantitative detections assessment (*Continued*)

Location With Max Detection	Analyte	CAS Number	*n*	Qualitative Frequency of Detection %[a]	Quantitative Frequency of Detection %[a]	Max Source Water Conc *ng/L*[a]	Max WWTP-Influenced Conc *ng/L*[b]	Source as % of WWTP Impacted Conc
	PFOA	335-67-1	25	100	76	112.0	—	—
	PFDA	335-76-2	25	92	60	31.1	—	—
	PFUnDA	2058-94-8	25	36	32	2.9	—	—
	PFDoDA	307-55-1	25	20	8	0.3	—	—
	Carisoprodol	78-44-4	25	16	4	5.0	155.9	3.2
	Fluconazole	86386-73-4	25	8	4	33.7	232.4	14.5
	Meprobamate	57-53-4	25	32	4	14.2	405.9	3.5
	Sulfadimethoxine	122-11-2	25	8	4	7.0	1,800.0	0.4
DWTP 24	PFBS	375-73-5	25	100	96	11.1	—	—
	PFHxS	355-46-4	25	92	92	44.8	—	—
	Tributyl phosphate	126-73-8	25	8	4	87.0	503.0	17.3
DWTP 26	Bupropion	34841-39-9	25	20	20	9.4	159.6	5.9
	Cholesterol	57-88-5	25	24	4	200.0	3,170.0	6.3
DWTP 27	Methyl-1H-benzotriazole	136-85-6	25	48	44	1,199.9	921.0	130.3
	Sulfamethoxazole	723-46-6	25	60	40	161.1	1,500.0	10.7
	Pseudoephederine	90-82-4	25	24	20	4.5	89.0	5.0
	Desmethyldiltiazem	85100-17-0	25	8	8	6.0	19.8	30.3
	Methocarbamol	532-03-6	25	36	8	32.3	2,627.3	1.2
	Hydrocodone	125-29-1	25	4	4	8.1	45.8	17.7
	Ranitidine	66357-35-5	25	4	4	13.1	313.9	4.2
DWTP 29	Lidocaine	137-58-6	25	20	8	29.7	408.8	7.3

[a]Source water concentrations are taken from Glassmeyer et al. 2017.

[b]Wastewater-influenced concentrations are taken from Bradley et al. 2017.

Conc—concentration, DWTP—drinking water treatment plant, WWTP—wastewater treatment plant

maximum concentrations for 40 CECs also measured in Bradley et al. (2017) are listed in Table 5, along with the calculated relationship between the source and wastewater-influenced concentrations. (An additional 80 analytes detected in Bradley et al. were also monitored in Glassmeyer et al. [2017], but they were not quantitatively detected in the source water samples and therefore excluded from the analysis.) The affected surface water sites sampled in Bradley et al. (2017) were chosen to reflect mixed-contaminant exposure profiles, including but not limited to wastewater effluent; they are not the same locations as Glassmeyer et al. (2017), but represent impacted locations. Of the 40 source water maximum concentrations from Glassmeyer et al. (2017), 34 were <20% of the more wastewater-influenced sample maximum reported in Bradley et al. (2017; Table 5). The source water concentrations of cotinine, diltiazem, and desmethyl diltiazem were all between 25% and about 30% of the wastewater-influenced maximums, while verapamil, amitriptyline, and methyl-1H-benzotriazole were found in source waters at concentrations greater than the wastewater-influenced locations (355, 118, and 130% relative concentration; Table 5). The fact that 114 out of 120 contaminant pairs monitored in both studies were substantially lower in the drinking water source waters demonstrates the beneficial effect that dilution and other natural attenuation processes have on aquatic CEC concentrations. However, the six compounds with >25% relative concentration, and particularly the three components with higher concentrations (verapamil, amitriptyline, and methyl-1H-benzotriazole), illustrate the need for DWTPs to estimate and assess the potential for WWTP influence on the chemical contaminant composition of their source water. DRINCS can identify situations in which potential exists for CECs in DWTP influents, and assist with understanding the potential seasonal variability as a function of streamflow (Rice & Westerhoff 2017, 2015). For DWTPs with higher levels of DFR under average flow (>1–5%), the frequency of CEC detection and CEC concentrations should be high enough to detect by modern analytical methods. Lower DFR values may have CECs that occur below current analytical detection limits (Rice et al. 2015). Thus, DRINCS emerges as a potentially useful tool to identify DWTPs at higher risk for CEC occurrence, where subsequent monitoring could be focused.

CONCLUSIONS

The variability in CEC detection at a particular DWTP intake depends on many factors including streamflow, type of treatment processes used at any upstream WWTP, WWTP discharge flow rates, travel distance, water quality within the receiving waters, and so on. As indicated in the prior study in which the CEC occurrence data were collected (Glassmeyer et al. 2017), the conclusion noted that samples collected at a single point in time make up a snapshot of occurrence, and future studies would benefit from more detailed and focused time series sample collection designs that better capture temporal variations. The general comparison of DRINCS and the "snapshot" of CEC occurrence data compared here advances the validity of using DRINCS as a tool to identify locations of DWTPs for future sampling and treatment technology testing. Before development and simulation of the DRINCS model (Rice & Westerhoff 2015), the only other available nationwide documentation linking drinking water sources to wastewater percentage was several decades old (Swayne et al. 1980). Levels of DFR from DRINCS were previously compared with the potential occurrence of Unregulated Contaminant Monitoring Rule CECs (Rice & Westerhoff 2015), which included only a few wastewater indicator compounds. However, this paper demonstrates, for the first time, the ability of DRINCS for a much broader range of CECs of wastewater origin and considers distances between WWTP discharges and DWTP intakes. Databases linked with DRINCS include populations served and type of unit processes at WWTPs and DWTPs; in addition, DRINCS is able to calculate number, size, and proximity of WWTP discharges into surface waters upstream of DWTP intakes. Queries could be made that include some of the factors described herein that would affect CEC occurrence. Although the comparison of model and field results in this study indicates the general validity of the DRINCS model, the data also suggest that predictive capabilities could be enhanced by closer proximity of instream flow information, such as that provided by streamgages near DWTP intakes, to more accurately measure DFR. Ongoing improvements in chemical analytical capabilities and expansion of the range of CECs routinely determined will also serve to better anchor model predictions with observed ambient source water conditions.

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ENDNOTE

[1]ArcGIS 10, Environmental Systems Research Institute, Redlands, Calif.

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